

Form PTO-1300 (Rev. 12-29-99)		US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NO. H 3891 PCT/US	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				U.S. APPLICATION NO. (If known, enter in box 16) <div style="font-size: 1.5em; font-weight: bold; text-align: center;">097/890295</div>	
INTERNATIONAL APPLICATION NO. PCT/EP00/00467		INTERNATIONAL FILING DATE January 22, 2000		PRIORITY DATE CLAIMED January 30, 1999	
TITLE OF INVENTION USE OF FATTY ACID ALKANOLAMINE ESTERS AS ANTISTATICS					
APPLICANT(S) FOR DO/EO/US Sergio Milan					
Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:					
<ol style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1). 4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)). <ol style="list-style-type: none"> a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). 7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). b. <input type="checkbox"/> have been transmitted by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input checked="" type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). 10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). 					
Items 11. to 16. below concern other document(s) or information included:					
<ol style="list-style-type: none"> 11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 12. <input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. <input checked="" type="checkbox"/> A FIRST preliminary amendment <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 14. <input type="checkbox"/> A substitute specification. 15. <input type="checkbox"/> A change of power of attorney and/or address letter. 16. <input type="checkbox"/> Other items or information: 					
"Express Mail Post Office to Addressee" service Mailing Label Number <u>EL541613721US</u>					

U.S. Application No. <u>09/7890293</u> If known, see 37 CFR 1.53	INTERNATIONAL FILING NO. PCT/EP00/00467	ATTORNEY'S DOCKET NUMBER H 3891 PCT/US																								
17. The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)): Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO..... \$1,000.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO..... \$860.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$710.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)..... \$100.00		<table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <th style="text-align: left;">CALCULATIONS</th> <th style="text-align: left;">PTO USE ONLY</th> </tr> <tr> <td colspan="2" style="height: 100px;"> <div style="display: flex; justify-content: space-between;"> <div> ENTER APPROPRIATE BASIC FEE AMOUNT = Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date 37 (CFR 1.492(e)). <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 30%;">CLAIMS</th> <th style="width: 20%;">NUMBER FILED</th> <th style="width: 20%;">NUMBER EXTRA</th> <th style="width: 30%;">RATE</th> </tr> </thead> <tbody> <tr> <td>Total Claims</td> <td>8 - 20 =</td> <td>0</td> <td>0 X \$18.00</td> </tr> <tr> <td>Independent Claims</td> <td>5 - 3 =</td> <td>2</td> <td>2 X \$80.00</td> </tr> <tr> <td colspan="3">Multiple dependent claims (s)/(if applicable)</td> <td>0 + \$270.00</td> </tr> <tr> <td colspan="3">TOTAL OF ABOVE CALCULATIONS</td> <td>=</td> </tr> </tbody> </table> </div> <div> \$ 860 \$ 0 \$ 0 \$ 1020 \$ 0 \$ 0 \$ 1020 \$ 0 \$ 1020 \$ 0 \$ 1020 </div> </div> </td> </tr> </table>	CALCULATIONS	PTO USE ONLY	<div style="display: flex; justify-content: space-between;"> <div> ENTER APPROPRIATE BASIC FEE AMOUNT = Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date 37 (CFR 1.492(e)). <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 30%;">CLAIMS</th> <th style="width: 20%;">NUMBER FILED</th> <th style="width: 20%;">NUMBER EXTRA</th> <th style="width: 30%;">RATE</th> </tr> </thead> <tbody> <tr> <td>Total Claims</td> <td>8 - 20 =</td> <td>0</td> <td>0 X \$18.00</td> </tr> <tr> <td>Independent Claims</td> <td>5 - 3 =</td> <td>2</td> <td>2 X \$80.00</td> </tr> <tr> <td colspan="3">Multiple dependent claims (s)/(if applicable)</td> <td>0 + \$270.00</td> </tr> <tr> <td colspan="3">TOTAL OF ABOVE CALCULATIONS</td> <td>=</td> </tr> </tbody> </table> </div> <div> \$ 860 \$ 0 \$ 0 \$ 1020 \$ 0 \$ 0 \$ 1020 \$ 0 \$ 1020 \$ 0 \$ 1020 </div> </div>		CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	Total Claims	8 - 20 =	0	0 X \$18.00	Independent Claims	5 - 3 =	2	2 X \$80.00	Multiple dependent claims (s)/(if applicable)			0 + \$270.00	TOTAL OF ABOVE CALCULATIONS			=
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Reduction of 1/2 for filing by small entity, if applicable. A Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28). SUBTOTAL = \$ 1020 Processing fee of \$130.00 for furnishing the English translation later the <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)). + \$ 0 TOTAL NATIONAL FEE = \$ 1020 Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property + \$ 0 TOTAL FEES ENCLOSED = \$ 1020		<table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 60%;">Amount to be refunded:</td> <td style="width: 40%;">\$-----</td> </tr> <tr> <td>charged:</td> <td>\$1020.00</td> </tr> </table>	Amount to be refunded:	\$-----	charged:	\$1020.00																				
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a. <input type="checkbox"/> A check in the amount of \$_____ to cover the above fees is enclosed. b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. <u>50-1177</u> in the amount of <u>\$1020.00</u> to cover the above fees. A triplicate copy of this sheet is enclosed. Order No. <u>01-0453</u> . c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>50-1177</u> . A triplicate copy of this sheet is enclosed. NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.																										
SEND ALL CORRESPONDENCE TO: Cognis Corporation, Law Dept. 2500 Renaissance Blvd., Suite 200 Gulph Mills, PA 19406																										
John E. Drach NAME ATTORNEY FOR APPLICANT 32,891 REGISTRATION NUMBER		 SIGNATURE:																								

"Express Mail" mailing label number EL541613721US.

PATENT
Docket No. H 3891 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

RE: PCT/EP00/00467
International Filing Date: January 22, 2000
Priority Date Claimed: January 30, 1999
Applicant: Sergio Milan
Title: USE OF FATTY ACID ALKANOLAMINE ESTERS AS
ANTISTATICS
Applicants' Reference: H 3891 PCT/US

PRELIMINARY AMENDMENT

Commissioner for Patents
Box PCT
Washington, DC 20231

ATTN: DO/EO/US

Sir:

Before examination, in the national stage for the United States, of the above-captioned application under the Patent Convention Treaty, please amend as follows the translation supplied herewith of the application:

In the Specification:

Please delete all text above line 7, of page 1, and replace the deleted matter with the following new section headings and new paragraph:

--TITLE OF THE INVENTION

Use of Fatty Acid Alkanolamine Esters as Antistatics

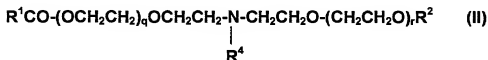
BACKGROUND OF THE INVENTION

This invention relates generally to thermoplastics and more particularly to the use of selected nitrogen-containing surfactants as antistatic additives.--

Please replace the paragraph beginning on page 4, line 8 and ending at page 4, line 19, with the following rewritten paragraph:

**Preliminary Amendment of U.S. National Stage for International
Application PCT/EP00/00467 filed January 22, 2000**

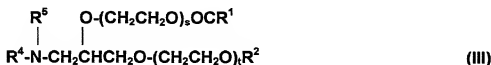
—Besides the fatty acid triethanolamine esters, other suitable antistatic agents are esters of fatty acids with diethanolalkyamines corresponding to formula (II):



in which R¹CO is an acyl group containing 6 to 22 carbon atoms, R² is hydrogen or has the same meaning as R¹CO, R⁴ is an alkyl group containing 1 to 4 carbon atoms and q and r together stand for 0 or numbers of 1 to 12.—

Please replace the paragraph beginning on page 4, line 20 and ending at page 4, line 32, with the following rewritten paragraph:

—Finally, a third group of suitable trialkanolamine esters are the esters of fatty acids with 1,2-dihydroxypropyl dialkylamines corresponding to formula (III):



in which R¹CO is an acyl group containing 6 to 22 carbon atoms, R² is hydrogen or has the same meaning as R¹CO, R⁴ and R⁵ independently of one another are alkyl groups containing 1 to 4 carbon atoms and s and t together stand for 0 or numbers of 1 to 12. So far as the choice of the preferred fatty acids and the optimal degree of esterification are concerned,—

Please replace the paragraph beginning on page 5, line 5 and ending at page 5, line 32, with the following rewritten paragraph:

—In another preferred embodiment of the invention, the fatty acid alkanolamine esters are used together with lubricants of the partial glyceride type which produce a synergistic improvement in color stability. Partial

**Preliminary Amendment of U.S. National Stage for International
Application PCT/EP00/00467 filed January 22, 2000**

glycerides, i.e. monoglycerides, diglycerides and technical mixtures thereof may still contain small quantities of triglycerides from their production. The partial glycerides preferably correspond to formula (IV):



in which R^6CO is a linear or branched, saturated and/or unsaturated acyl group containing 6 to 22 and preferably 12 to 18 carbon atoms, R^7 and R^8 independently of one another have the same meaning as R^6CO or represent OH and the sum ($v+w+x$) is 0 or a number of 1 to 100 and preferably 5 to 25, with the proviso that at least one of the two substituents R^6 and R^7 represents OH.

Typical examples are mono- and/or diglycerides based on caproic acid, caprylic acid, 2-ethylhexanoic acid, capric acid, lauric acid, isotridecanoic acid, myristic acid, palmitic acid, palmitoleic acid, stearic acid, isostearic acid, oleic acid, elaidic acid, petroselinic acid, linoleic acid, linolenic acid, elaeostearic acid, arachic acid, gadoleic acid, behenic acid and erucic acid and technical mixtures thereof. Technical lauric acid glycerides, palmitic acid glycerides, stearic acid glycerides, isostearic acid glycerides, oleic acid glycerides, behenic acid glycerides and/or erucic acid glycerides which have a monoglyceride content of 50 to 95% by weight and preferably 60 to 90% by weight are preferably used. The ratio by weight—

In the claims:

Please cancel claims 1-9.

Please add the following new claims 10-17.

10. (New) A method of imparting antistatic properties to a thermoplastic comprising contacting a thermoplastic with from about 0.5 to about 5 parts by weight of an antistatic agent selected from the group consisting of (1) a compound of the formula (IV):

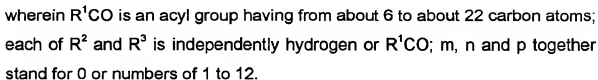


wherein R^6CO is a linear or branched, saturated and/or unsaturated acyl group having from 6 to 22 carbon atoms; each of R^7 and R^8 is R^6CO or OH with the proviso that at least one of R^6 and R^7 is OH; each of m, n, and p is a number for 0 to 100 such that the sum of v+w+x has a value of from 0 to 100; (2) a compound of the formula (V):



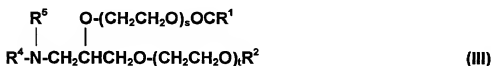
wherein R^9CO is a linear or branched, saturated or unsaturated acyl group having from 6 to 22 carbon atoms and combinations of (1) and (2).

11. (New) The method of claim 10 wherein the number of carbon atoms in the R^6CO group is from about 12 to about 18.
12. (New) The method of claim 10 wherein the number of carbon atoms in the R^9CO group is from about 12 to about 18.
13. (New) The method of claim 10 wherein when compounds (IV) and (V) are present together, the weight ratio of (IV) to (V) is from about 90:10 to about 10:90.
14. (New) A method of imparting antistatic properties to a thermoplastic comprising contacting a thermoplastic with from about 0.5 to about 5 parts by weight of an antistatic agent of the formula (I):


$$\text{R}^1\text{CO}-(\text{OCH}_2\text{CH}_2)_q\text{OCH}_2\text{CH}_2-\underset{\text{R}^4}{\text{N}}-\text{CH}_2\text{CH}_2\text{O}-(\text{CH}_2\text{CH}_2\text{O})_r\text{R}^2 \quad (\text{II})$$

wherein R¹CO is an acyl group having from about 6 to about 22 carbon atoms, R² is hydrogen or R¹CO; R⁴ is an alkyl group having from 1 to about 4 carbon atoms and q and r together stand for 0 or numbers of 1 to 12.

16. (New) A method of imparting antistatic properties to a thermoplastic comprising contacting a thermoplastic with from about 0.5 to about 5 parts by weight of an antistatic agent of the formula (III):



wherein R¹CO is an acyl group having from about 6 to about 22 carbon atoms; R² is hydrogen or R¹CO, each of R⁴ and R⁵ is independently an alkyl group having 1 to about 4 carbon atoms and s and t together stand for 0 or numbers of 1 to 12.

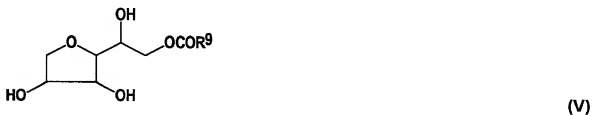
17. (New) A composition comprising (A) an antistatic agent selected from the

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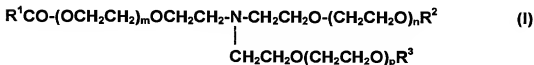
group consisting of (1) a compound of the formula (IV):



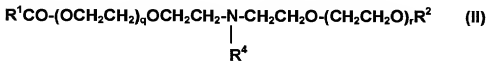
wherein R^6CO is a linear or branched, saturated and/or unsaturated acyl group having from 6 to 22 carbon atoms; each of R^7 and R^8 is R^6CO or OH with the proviso that at least one of R^6 and R^7 is OH; each of m, n, and p is a number for 0 to 100 such that the sum of $v+w+x$ has a value of from 0 to 100; (2) a compound of the formula (V):



wherein R^9CO is a linear or branched, saturated or unsaturated acyl group having from 6 to 22 carbon atoms; (3) a compound of the formula (I):



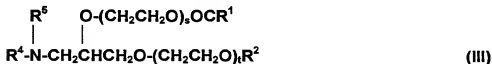
wherein R^1CO is an acyl group having from about 6 to about 22 carbon atoms; each of R^2 and R^3 is independently hydrogen or R^1CO ; m, n and p together stand for 0 or numbers of 1 to 12; (4) a compound of the formula (II):



wherein R^1CO is an acyl group having from about 6 to about 22 carbon atoms, R^2 is hydrogen or R^1CO ; R^4 is an alkyl group having from 1 to about 4 carbon

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Application PCT/EP00/00467 filed January 22, 2000**

atoms and q and r together stand for 0 or numbers of 1 to 12; (5) a compound of the formula (III):



wherein R¹CO is an acyl group having from about 6 to about 22 carbon atoms; R² is hydrogen or R¹CO, each of R⁴ and R⁵ is independently an alkyl group having 1 to about 4 carbon atoms and s and t together stand for 0 or numbers of 1 to 12 and, (B) a thermoplastic selected from the group consisting of low-density polyethylene, high-density polyethylene, polypropylene, polystyrene, a vinyl polymer, a polyamide, a polyester, a polyacetal, a polycarbonate and a polyurethane.

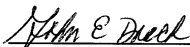
REMARKS

Claims 10-17 are currently pending in the instant application.

The Specification has been amended to include the preferred section headings pursuant to 37 C.F.R. §1.77 to remove possible ambiguities associated with the use of the same subscript letters which stand for the degrees of ethoxylation in formulas II, III, and IV. It is submitted that the amendments to the Specification made herein introduce no new matter. Their entry is therefore proper and respectfully requested. An Abstract of the Disclosure has been added on a separate sheet following the claims.

Original claims 1-9 have been canceled and replaced with new claims 10-17 solely for the purpose of improving clarity and grammar, which may suffer in translation, and not for any reason related to the statutory requirements for a patent. New claims 10-17 have not been added in response to any rejection, or in anticipation of any rejection related to the statutory requirements for a patent. Applicants respectfully submit that the scope of new claims 10-17 corresponds to the scope of original claims 1-9 and that new claims 10-17 are no narrower than original claims 1-9. Furthermore, although a moot point in view of their cancellation, Applicants respectfully submit that original claims 1-9 satisfied the requirements of 35 U.S.C. §112, as filed. New claims 10-17 are supported by the specification and no new matter has been introduced. Entry is therefore proper and respectfully requested. Prompt examination of the instant application in view of the amendments made herein is respectfully requested.

Respectfully submitted,



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(Reg. No. 32,891)
Attorney for Applicants
(610) 278-4925

Cognis Corporation, Patent Dept.
2500 Renaissance Blvd., Suite 200
Gulph Mills, PA 19406

JED/ras

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Use of Fatty Acid Alkanolamine Esters as Antistatics

Field of the Invention

This invention relates generally to thermoplastics and more particularly to the use of selected nitrogen-containing surfactants as antistatic additives.

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Prior Art

Thermoplastics, preferably PVC, are used for the production of a number of articles of everyday life, of which films are among the most well-known. Like most plastics, thermoplastics are pronounced insulators by virtue of their chemical constitution. Unfortunately, a disadvantage is that thermoplastics readily develop electrostatic charges and, once applied, charges cannot be dissipated quickly enough on account of the low surface conductivity. In practice, the electrostatic charging of plastics can be troublesome and can also give rise to serious dangers. These include above all:

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- heavy soiling of plastic surfaces,
- production stoppages through the blocking of film webs and
- sparking through intensive charging with subsequent ignition of dust/air mixtures.

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In order to solve the problem of static charging, antistatic agents are generally added to the thermoplastics to dissipate the charges from the surface. Examples of internal antistatic agents, i.e. substances which are added to the polymer melt before or during processing, are anionic, nonionic or cationic surfactants. A relevant overview was published by S.

25

Riethmayer in **Gummi, Asbest, Kunstst.**, 26, pp. 76-88, 182-184, 298-

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308, 419,429, 507-512 (1973).

Japanese patent application **JP 94/226266** (Henkel) describes antistatic agents for PVC in the form of mixtures of complex esters, alkyl benzenesulfonates and alkyl sulfates. Polyol complex esters are known for the same purpose from German patent application **DE-A1 4304468** (Henkel). Finally, antistatic agents for PVC in the form of mixtures of anionic surfactants (secondary alkane sulfonates) and nonionic surfactants (amine polyglycol ethers) are commercially obtainable under the name of "Dehydant 80-X" from Henkel KGaA.

However, the properties of known antistatic agents are still in need of improvement. They either fail to reduce the surface charge of the thermoplastics sufficiently quickly to the required level or, despite satisfactory antistatic properties, they lead to unwanted clouding in the films.

Accordingly, the problem addressed by the present invention was to finish thermoplastics in general and polyvinyl chloride and polyolefins in particular in such a way that, on the one hand, electrostatic charging would be significantly reduced and, on the other hand, transparent films permanently protected against clouding would be obtained.

Description of the Invention

The present invention relates to the use of fatty acid alkanolamine esters as antistatic agents for thermoplastics, more particularly for polyvinyl chloride and polyolefins.

It has surprisingly been found that fatty acid alkanolamine esters not only provide thermoplastics with excellent antistatic properties, they also lead to transparent films with hardly any tendency towards clouding, even in the event of prolonged storage.

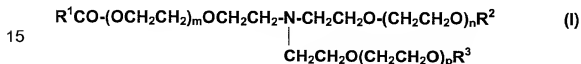
Thermoplastics

In the context of the invention, the antistatic finish may be applied to

thermoplastics in general. Typical examples are polyolefins, such as low-density and high-density polyethylene, polypropylene, polystyrene, vinyl polymers, polyamides, polyesters, polyacetals, polycarbonates and polyurethanes. However, the antistatic agents used in accordance with the invention are preferably incorporated in polyvinyl chlorides with K values in the range from 30 to 80.

Fatty acid alkanolamine esters

The fatty acid alkanolamine esters are known substances. In a first and also preferred embodiment of the invention, the antistatic agents used are fatty acid triethanolamine esters which preferably correspond to formula (I):



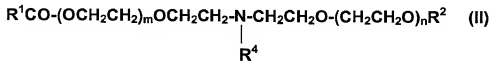
in which R^1CO is an acyl group containing 6 to 22 carbon atoms, R^2 and R^3 independently of one another represent hydrogen or have the same meaning as R^1CO and m, n and p together stand for 0 or numbers of 1 to 12. Typical examples of fatty acid triethanolamine esters which may be used in accordance with the present invention are products based on caproic acid, caprylic acid, capric acid, lauric acid, myristic acid, palmitic acid, isostearic acid, stearic acid, oleic acid, elaidic acid, arachic acid, behenic acid and erucic acid and the technical mixtures thereof obtained, for example, in the pressure hydrolysis of natural fats and oils. Technical $\text{C}_{12/18}$ coconut fatty acids and, in particular, partly hydrogenated $\text{C}_{16/18}$ tallow or palm oil fatty acids and $\text{C}_{16/18}$ fatty acid cuts rich in elaidic acid are preferably used. To produce the esters, the fatty acids and the triethanolamine may be used in a molar ratio of 1.1:1 to 3:1. With the performance properties of the esters in mind, a ratio of 1.2:1 to 2.2:1 and preferably 1.5:1 to 1.9:1 has proved to be particularly advantageous. The

preferred fatty acid triethanolamine esters are technical mixtures of mono-, di- and triesters with an average degree of esterification of 1.5 to 1.9 and are derived from technical C_{18/18} tallow or palm oil fatty acid (iodine value 0 to 40). In performance terms, fatty acid triethanolamine ester salts

5 corresponding to formula (I), in which R¹CO is an acyl group containing 16 to 18 carbon atoms, R² has the same meaning as R¹CO, R³ is hydrogen and m, n and p stand for 0 have proved to be particularly advantageous.

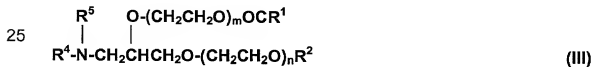
Besides the fatty acid triethanolamine esters, other suitable antistatic agents are esters of fatty acids with diethanolalkylamines corresponding to

10 formula (II):



15 in which R¹CO is an acyl group containing 6 to 22 carbon atoms, R² is hydrogen or has the same meaning as R¹CO, R⁴ is an alkyl group containing 1 to 4 carbon atoms and m and n together stand for 0 or numbers of 1 to 12.

20 Finally, a third group of suitable trialkanolamine esters are the esters of fatty acids with 1,2-dihydroxypropyl dialkylamines corresponding to formula (III):



25 in which R¹CO is an acyl group containing 6 to 22 carbon atoms, R² is hydrogen or has the same meaning as R¹CO, R⁴ and R⁵ independently of one another are alkyl groups containing 1 to 4 carbon atoms and m and n together stand for 0 or numbers of 1 to 12. So far as the choice of the preferred fatty acids and the optimal degree of esterification are concerned,

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Partial glycerides

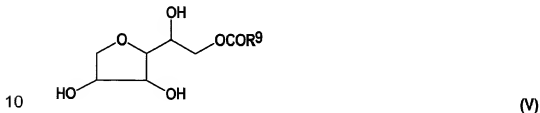
$$\begin{array}{c} \text{CH}_2\text{O}(\text{CH}_2\text{CH}_2\text{O})_m\text{COR}^6 \\ | \\ \text{CHO}(\text{CH}_2\text{CH}_2\text{O})_n\text{R}^7 \\ | \\ \text{CH}_2\text{O}(\text{CH}_2\text{CH}_2\text{O})_p\text{R}^8 \end{array} \quad (\text{IV})$$

in which R⁶CO is a linear or branched, saturated and/or unsaturated acyl group containing 6 to 22 and preferably 12 to 18 carbon atoms, R⁷ and R⁸ independently of one another have the same meaning as R⁶CO or represent OH and the sum (m+n+p) is 0 or a number of 1 to 100 and preferably 5 to 25, with the proviso that at least one of the two substituents R⁶ and R⁷ represents OH. Typical examples are mono- and/or diglycerides based on caproic acid, caprylic acid, 2-ethylhexanoic acid, capric acid, lauric acid, isotridecanoic acid, myristic acid, palmitic acid, palmitoleic acid, stearic acid, isostearic acid, oleic acid, elaidic acid, petroselic acid, linoleic acid, linolenic acid, elaeostearic acid, arachic acid, gadoleic acid, behenic acid and erucic acid and technical mixtures thereof. Technical lauric acid glycerides, palmitic acid glycerides, stearic acid glycerides, isostearic acid glycerides, oleic acid glycerides, behenic acid glycerides and/or erucic acid glycerides which have a monoglyceride content of 50 to 95% by weight and preferably 60 to 90% by weight are preferably used. The ratio by weight

between the trialkanolamine esters and the partial glycerides may be in the range from 90:10 to 10:90 and is preferably in the range from 75:25 to 25:75 and more particularly in the range from 60:40 to 40:60.

5 Sorbitan esters

In another preferred embodiment of the invention, the fatty acid alkanolamine esters are used together with lubricants of the sorbitan ester type which also produce a synergistic improvement in color stability. Sorbitan esters preferably correspond to formula (V):



15 in which R^9CO represents linear or branched, saturated or unsaturated acyl groups containing 6 to 22 and preferably 12 to 18 carbon atoms. Although formula (I) only represents sorbitan monoesters, sorbitan di-, sesqui- and tri-esters and mixtures thereof are also suitable. Typical examples are mono-, sesqui-, di- and/or triesters of sorbitan with caproic acid, caprylic acid, 2-ethyl hexanoic acid, capric acid, lauric acid, isotridecanoic acid, myristic acid, palmitic acid, palmitoleic acid, stearic acid, isostearic acid, oleic acid, elaidic acid, petroselic acid, linoleic acid, linolenic acid, 20 elaeostearic acid, arachic acid, gadoleic acid, behenic acid and erucic acid and technical mixtures thereof. The ratio by weight between the trialkanolamine esters and the sorbitan esters may be in the range from 90:10 to 10:90 and is preferably in the range from 75:25 to 25:75 and more particularly in the range from 60:40 to 40:60.

25

Internal and external application

The thermoplastics are antistatically finished, for example, by mixing

the powdered or granulated polymers with the additives and intensively homogenizing the resulting mixture. This may be done by individually adding the additives, i.e. for example flow improvers, stabilizers, lubricants, antistatic agents, plasticizers, etc. However, a so-called masterbatch, i.e. a homogeneous mixture of all additives, may also be prepared and then mixed with the plastic. It is advisable to carry out the mixing step in the presence of heat, preferably above the softening point of the thermoplastic, and then immediately to process the additive-containing mixture, i.e. for example by extrusion, injection molding, calendaring, rolling, blow molding, stretch forming and the like. Besides being used as internal antistatic agents, the fatty acid alkanolamine esters may also be used as external antistatic agents, i.e. the esters are applied to the surface from a solution in water or an alcohol.

15 Auxiliaries and additives

The fatty acid alkanolamine esters may be used together with other auxiliaries and additives for the production and processing of thermoplastics such as, for example, flow improvers and impact modifiers, lubricants, stabilizers, plasticizers, co-antistatics, fillers, pigments and the like.

Examples of suitable **impact modifiers** are ethylene/vinyl acetate copolymers as graft base, ethylene/vinyl acetate/vinyl chloride graft polymers, polyacrylate/vinyl chloride graft polymers, acrylate/methyl methacrylate graft polymers, chlorinated polyethylene, methyl methacrylate/butadiene/styrene graft polymer and acrylonitrile/butadiene/styrene terpolymer (the last two for internal application).

Typical lubricants include, for example, hydrocarbons (paraffin oils, natural paraffins, synthetic paraffins, low-density and high-density polyethylene waxes, polypropylene waxes), alcohols (cetyl alcohol, stearyl alcohol, tallow fatty alcohol), ketones (stearone), carboxylic acids (lauric

acid, myristic acid, palmitic acid, stearic acid, hydroxystearic acid, tallow fatty acid, arachic acid, behenic acid, montanic acid, oxidized polyethylene waxes), metal salts of carboxylic acids (calcium stearate, zinc stearate, lead stearate, calcium montanate, calcium salts of oxidized polyethylene waxes or synthetic wax acids), carboxylic acid amides (oleic acid amide, erucic acid amide, stearic acid amide, ethylene distearoyl diamide), carboxylic acid esters (ethyl stearate, n-butyl stearate, isobutyl stearate, isooctyl stearate, isotridecyl stearate, cetyl palmitate, cetyl stearate, ethylene glycol monostearate, glycerol monooleate, glycerol monoricinoleate, glycerol monostearate, glycerol mono-12-hydroxystearate, glycerol tristearate, glycerol tri-12-hydroxystearate, glycerol tribehenate, glycerol trimontanate, pentaerythritol tetrastearate, pentaerythritol tetrabehenate, mixed esters of adipic acid, pentaerythritol and stearic acid, montanic acid esters, partly saponified montanic acid esters). In one preferred embodiment of the invention, the esterquats are used together with lubricants of the fatty acid, fatty alcohol and/or partial glyceride type, the ratio by weight of the components being in the range from 90:10 to 60:40. The mixtures have the advantage of a very positive effect on the color stability of the plastics.

The **stabilizers** are divided into UV absorbers (hydroxybenzophenones, hydroxyphenyl benzotriazoles, cinnamic acid esters, oxalanilides), quenchers (essentially nickel complexes), hydroperoxide decomposers (thiocarbamates, thiophosphates, thiobisphenolates) and radical scavengers (sterically hindered amines).

The following substances - which are used in particular for stabilizing polyvinyl chloride - are mentioned as representative of the large number of stabilizers: 2-hydroxy-4-n-octoxy benzophenone, 2-(2'-hydroxy-5'-methylphenyl)-benzotriazole, 2-(2'-hydroxy-3',5'-di-tert.butylphenyl)-benzotriazole, 2,2'-dihydroxybenzophenone, 2-hydroxy-4-methoxybenzophenone, N,N'-(2-ethyl-2'-ethoxyphenyl)-oxalic acid amide, 2-carbomethoxy-

4'-methoxycinnamic acid methyl ester, 2-cyano-3-methyl-4'-methoxycinnamic acid methyl ester, sebacic acid bis-2,2,6,6-tetramethyl-4-piperidyl ester, organotin carboxylates. β -keto compounds such as, for example, β -diketones and β -ketocarboxylic acids are also suitable.

- 5 Examples of suitable **plasticizers** are phthalic acid esters (dimethyl phthalate, diethyl phthalate, dibutyl phthalate, dihexyl phthalate, di-2-ethyl hexyl phthalate, di-n-octyl phthalate, di-i-octylphthalate, di-i-nonyl phthalate, di-i-decyl phthalate, di-i-tridecyl phthalate, dicyclohexyl phthalate, dimethyl cyclohexyl phthalate, dimethyl glycol phthalate, dibutyl glycol phthalate,
- 10 benzyl butyl phthalate, diphenyl phthalate), phosphoric acid esters (tributyl phosphate, tri-2-ethylbutyl phosphate, tri-2-ethyl hexyl phosphate, trichloroethyl phosphate, 2-ethyl hexyl diphenyl phosphate, cresyl diphenyl phosphate, triphenyl phosphate, tricresyl phosphate, trixylenyl phosphate), esters of adipic, azelaic and sebacic acid (di-2-ethyl hexyl adipate, di-i-octyl
- 15 adipate, di-i-nonyl adipate, di-i-decyl adipate, benzyl butyl adipate, benzyl octyl adipate, di-2-ethyl hexyl azelate, di-2-ethyl hexyl sebacate, di-i-decyl sebacate), fatty acid esters, epoxidized fatty acid esters, citric acid esters, esters of acetic, propionic and butyric acid, esters of ethyl butyric and ethyl hexanoic acid, glycolic acid esters, polyesters, chloroparaffins,
- 20 hydrocarbons, benzoic acid esters, trimellitic acid esters, sulfonic acid esters and sulfamides, alcohols, ethers and ketones and abietic acid esters.

- Suitable **fillers** are carbonates (calcium carbonate, dolomite), silicates (talcum, asbestos, kaolin, mica), silicon dioxide, aluminium
- 25 hydroxide, carbon black, organic substances (nutshells, wood meal, corn cobs), glass fibers, glass beads, hollow glass beads, carbon fibers, aramide fibers, whiskers and the like. Examples of suitable **pigments** are titanium dioxide, iron oxides, pigment blacks, chrome yellow pigments, molybdate red pigments, chrome oxide green pigments, mixed phase
- 30 pigments and cadmium pigments.

The auxiliaries may be added in total quantities - based on 100 parts by weight of thermoplastic - of 1 to 10 and preferably 4 to 8 parts by weight.

Examples

5

Examples 1 to 14, Comparison Examples C1 to C10. Polyvinyl chlorides and polyolefins were processed to films after the addition of various antistatic agents and auxiliaries [(tr) = transparent, (op) = opaque]. The surface of the films was then charged under defined conditions and the discharge time was measured. Antistatic activity is determined by multiplying the discharge time from 100 or 500 to 0 volt in h by ohms. The lower the value, the better the antistatic effect. The thermal stability of the polymers was defined by two parameters, i.e. the time interval (a) before the first sign of discoloration on exposure to heat and the time (b) elapsing before the film melts. The results are set out in Tables 1 and 2. Examples 1 to 14 correspond to the invention, Examples C1 to C10 are intended for comparison. Where fatty acid alkanolamine esters were used in accordance with the invention, optionally in admixture with other additives, transparent films with a good antistatic finish were obtained.

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Table 1.**Static charging of PVC (quantities in parts by weight)**

Component	C1	C2	C3	1	2	3	4
Polyvinyl chloride, K = 70	100	100	100	100	100	100	100
Diethyl phthalate	50	50	50	50	50	50	50
Epoxidized soybean oil	2	2	2	2	2	2	2
Calcium/zinc stabilizer	2	2	2	2	2	2	2
Glycerol monostearate	-	1	-	-	0.75	-	-
Sorbitan monopalmitate	-	-	1	-	-	0.75	0.25
Triethanolamine distearate	-	-	-	1	0.25	0.25	0.25
<i>Appearance of the PVC film</i>	op	tr	tr	tr	tr	tr	tr
<i>First yellowing [mins]</i>	20	20	20	20	20	20	20
<i>Static thermal stability [mins]</i>	50	50	50	>50	>50	>50	>50
<i>Residual surface charge ($10^{10} \Omega$)</i>							
- 100 V, 1d	143	7.2	7.5	5.9	1.3	1.3	1.3
- 500 V, 1d	83	6.1	6.0	4.3	1.5	1.5	1.5
- 100 V, 7d	22	0.9	0.8	0.2	0.2	0.2	0.2
- 500 V, 7d	19	0.6	0.5	0.2	0.2	0.2	0.2

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Table 1 continued.

Static charging of PVC (quantities in parts by weight)

Component	5	6	7	8	9	10	11
Polyvinyl chloride, K = 70	100	100	100	100	100	100	100
Diocetyl phthalate	50	50	50	50	50	50	50
Epoxidized soybean oil	2	2	2	2	2	2	2
Calcium/zinc stabilizer	2	2	2	2	2	2	2
Triethanolamine distearate	0.95	0.90	0.95	0.95	0.95	0.95	0.95
Pentaerythritol	0.05	0.10	-	-	-	-	-
Bisphenol A	-	-	0.05	-	-	-	-
Hydrotalcite	-	-	-	0.05	-	-	-
Butyl hydroxytoluene	-	-	-	-	0.05	-	-
Sorbitol	-	-	-	-	-	0.05	-
Di-trimethyl propane	-	-	-	-	-	-	0.05
Appearance of the PVC film	tr	tr	tr	tr	tr	tr	tr
First yellowing [mins]	50	60	30	30	45	30	20
Static thermal stability [mins]	60	60	80	70	70	60	50
Residual surface charge (10^8 C)							
- 100 V, 1d	10	18	200	260	480	950	190
- 500 V, 1d	13	24	120	2000	4000	5200	1200
- 100 V, 7d	60	24	180	85	32	23	30
- 500 V, 7d	44	24	180	64	38	22	26

Table 2.**Static charging of polyolefins (quantities in parts by weight)**

Component	12	C5	C6	C7	13	C8	C9	C10	14	C11	C12	C13
Low-density polyethylene	100	100	100	100	-	-	-	-	-	-	-	-
High-density polyethylene	-	-	-	-	100	100	100	100	-	-	-	-
Polypropylene	-	-	-	-	-	-	-	-	100	100	100	100
Triethanolamine distearate	0.03	-	-	-	0.03	-	-	-	0.03	-	-	-
Esterquat A*	-	0.03	-	-	-	0.03	-	-	-	0.03	-	-
Esterquat B**	-	-	0.03	-	-	-	0.03	-	-	-	0.03	-
Esterquat C***	-	-	-	0.15	-	-	-	0.15	-	-	-	0.15
Glycerol monostearate	0.12	0.12	0.12	-	0.12	0.12	0.12	-	0.12	0.12	0.12	-
Residual surface charge [$1 \times 10 \times 12$]												
-100 V, 3 d	11	12	12	18	16	18	17	17	16	18	16	18
-100 V, 8 d	9	10	10	17	16	17	15	14	14	14	14	18
-100 V, 30 d	9	10	11	13	12	14	13	13	12	13	14	15

*) Dehyquart AU 46: based on partly hydrogenated tallow fatty acid

**) Dehyquart AU 18: based on stearic acid

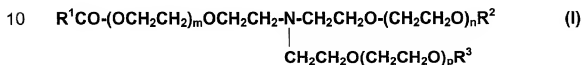
***) Dehyquart F30: mixture of (a) 30 parts esterquat based on partly hydrogenated palm oil fatty acid and (b) 70 parts stearyl alcohol

CLAIMS

1. The use of fatty acid alkanolamine esters as antistatic agents for thermoplastics.

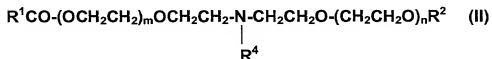
2. The use claimed in claim 1, characterized in that the fatty acid alkanolamine esters are used as antistatic agents for polyvinyl chlorides and polyolefins.

3. The use claimed in claims 1 and/or 2, characterized in that fatty acid alkanolamine esters corresponding to formula (I):



in which R^1CO is an acyl group containing 6 to 22 carbon atoms, R^2 and R^3 independently of one another represent hydrogen or have the same meaning as R^1CO and m , n and p together stand for 0 or numbers of 1 to 12, are used.

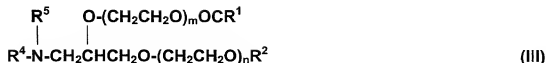
4. The use claimed in claims 1 and/or 2, characterized in that fatty acid alkanolamine esters corresponding to formula (II):



in which R^1CO is an acyl group containing 6 to 22 carbon atoms, R^2 is hydrogen or has the same meaning as R^1CO , R^4 is an alkyl group containing 1 to 4 carbon atoms and m and n together stand for 0 or numbers of 1 to 12,

are used.

5. The use claimed in claims 1 and/or 2, characterized in that fatty acid alkanolamine esters corresponding to formula (III):



5

in which R^1CO is an acyl group containing 6 to 22 carbon atoms, R^2 is hydrogen or has the same meaning as R^1CO , R^4 and R^5 independently of one another are alkyl groups containing 1 to 4 carbon atoms and m and n together stand for 0 or numbers of 1 to 12,

10 are used.

6. The use claimed in at least one of claims 1 to 5, characterized in that the fatty acid alkanolamine esters are used together with fatty acid partial glycerides corresponding to formula (IV):

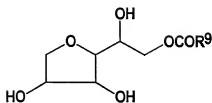


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in which R^6CO is a linear or branched, saturated and/or unsaturated acyl group containing 6 to 22 and preferably 12 to 18 carbon atoms, R^7 and R^8 independently of one another have the same meaning as R^6CO or represent OH and the sum $(m+n+p)$ is 0 or a number of 1 to 100, with the proviso that at least one of the two substituents R^6 and R^7 represents OH.

25

7. The use claimed in at least one of claims 1 to 6, characterized in that the fatty acid alkanolamine esters are used together with sorbitan esters corresponding to formula (V):



(V)

in which R^9CO represents linear or branched, saturated or unsaturated acyl groups containing 6 to 22 and preferably 12 to 18 carbon atoms.

5 8. The use claimed in at least one of claims 1 to 7, characterized in that the fatty acid alkanolamine esters and the partial glycerides or sorbitan esters are used in a ratio by weight of 90:10 to 10:90.

9. The use claimed in at least one of claims 1 to 8, characterized in that the fatty acid alkanolamine esters are used in quantities - based on 100
10 parts by weight of thermoplastic - of 0.5 to 5 parts by weight.

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ABSTRACT OF THE DISCLOSURE

Antistatic properties are imparted to thermoplastics by contacting the thermoplastics with fatty acid alkanolamine esters and/or fatty acid partial glycerides. Thermoplastics having these antistatic properties provided by the compounds according to the invention are transparent with little tendency toward clouding.

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0010PFTO Rev. 6/95 DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION <input type="checkbox"/> Declaration Submitted with Initial Filing OR <input checked="" type="checkbox"/> Declaration Submitted after Initial Filing	U.S. Department of Commerce Patent and Trademark Office	Attorney Docket Number	H 3891 PCT/US
		First Named Inventor	MILAN, Sergio
	COMPLETE IF KNOWN		
	Application Number		
	Filing Date		
	Group Art Unit		
		Examiner Name	

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

USE OF FATTY ACID ALKANOLAMINE ESTERS AS ANITSTATICS

the specification of which

(Title of the Invention)

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) **01/22/2000** as United States Application Number or PCT International

Application Number **PCT/EP00/00467** and was amended on (MM/DD/YYYY) (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §110(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached?	
				YES	NO
199 03 715.9	DE	01/30/1999	<input type="checkbox"/>	<input type="checkbox"/>	<input checked="" type="checkbox"/>

☐ Additional foreign application numbers are listed on a supplemental priority sheet attached hereto.

I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)	Additional provisional application numbers are listed on a supplemental priority sheet attached hereto.

Burden Hour Statement: This form is estimated to take .4 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Washington, DC 20231.

DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112.1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP00/00467	01/22/2000	

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

<input type="checkbox"/> Firm Name	<input type="checkbox"/> Customer Number	OR	<input type="checkbox"/> Label
<input checked="" type="checkbox"/> List Attorney(s) and/or agent(s) name and registration number below:			
Name	Registration Number	Name	Registration Number
John E. Drach	32,891	Aaron R. Ettelman	42,516
Steven J. Trzaska	36,296	Henry E. Millson, Jr.	18,980

☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

Please direct all correspondence to: ☒ Customer Number **23657** OR ☒ Fill in correspondence address below

Name	John E. Drach		
Address			
Address			
City	State	ZIP	
Country	Telephone	610-278-4925	Fax 610-278-6548

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this unsigned

Given Name	Sergio	Middle Initial		Family Name	Milan	Suffix e.g. Jr.	
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Inventor's Signature	<i>Sergio Milan</i>	Date	2001-06-25
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Residence: City	Barcelona	State		Country	Spain	Citizenship	Spain
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Post Office Address	Calle Lope de Vega, 290 3-1-B
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City	08018 Barcelona	State		Zip		Country	Spain	Applicant Authority	
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☐ Additional inventors are being named on supplemental sheet(s) attached hereto